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TOTAL EMISSIVITY OF HOT WATER VAPOR

I. THE HIGH-PRESSURE UPPER LIMIT

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ABSTRACT

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The [total emissivity of water vapor and the relative energy distributions in various broad-band spectral regions] have been calculated from experimental thin-gas spectral data at temperatures between 500° and 2200°K and optical path-lengths between 0.1 and 10,000 cm·atm. The calculation provides an upper limit to the emission which should be useful in heat transfer calculations and the design of thermal protection systems.

author

INTRODUCTION

The radiative heat transfer from the exhaust plume to the base of a launch vehicle which uses hydrogen and oxygen as propellants is due almost entirely to infrared radiation from hot water vapor. Radiation from water vapor is also an important part of the base heating of a hydrocarbon-oxygen vehicle. Previous investigations of the emission of hot H_2O have not included the conditions of temperature, pressure, and optical pathlength which are pertinent to calculations of radiative heating from the plumes of very large vehicles like the Saturn V or Nova. For this reason an investigation of the radiant emission from hot water has been initiated at the Space Science Laboratory of General Dynamics/Astronautics. This investigation is supported by the Aero-Astroynamics Laboratory (R-AERO-R) of the NASA's Marshall Space Flight Center. The investigation has as its goal the development of an ability to predict the transfer from an arbitrary volume of hot water vapor, under arbitrary (and non-uniform) conditions of temperature and pressure, to a nearby surface.

The present report is the first of a series of reports which will be issued as various steps are taken toward the goal. This report presents an evaluation of the total or "engineering" emissivity of a uniform sample of water vapor in the limit of very high pressure, where the spectral emissivity is described by Beer's Law with frequency-averaged empirical absorption coefficients. This is equivalent to the weak line approximation;

the calculation is exact at very low optical depths, and provides an absolute upper limit at large values of the optical depth.

Future reports will discuss the emissivity of Doppler-broadened H_2O (this is the low pressure limit to the emissivity) and the emissivity of H_2O when the spectral lines are collision-broadened but not smeared enough for the frequency-averaged model to be applied at large optical depths. Further experimental data, at long pathlengths, will be required for the collision-broadened case, and experimental work is presently in progress which will furnish these data.

THEORETICAL DEVELOPMENT

The spectral emissivity for radiation from homogeneous gases in a very narrow frequency range (ν to $\nu + \delta\nu$) along a given line of sight approaches the value given by

$$\epsilon(\nu, T, u) = 1 - \exp\{-uk(\nu, T)u\} \quad (1)$$

in the limit as $\delta\nu$ becomes vanishingly small. Equation (1), which is a statement of the well-known Beer-Lambert Law, relates the emissivity ϵ at given frequency ν , temperature T , and optical depth u , to the absorption coefficient k , a function of temperature and frequency for a particular gas. If calculations or measurements of k with sufficient spectral resolution are available, the total emission may in principle be calculated by the relation

$$N(T, u) = \int_0^{\infty} N_{\nu}^0(T) [1 - \exp\{-k(\nu, T)u\}] d\nu \quad (2)$$

Here N is the emission in watts/cm² and N_{ν}^0 is the Planck blackbody radiance function.

In general, however, gaseous spectral emissivities measured with even the highest-resolution spectrometers do not show the variation with optical depth predicted by Eq. (1). The emission measured by such an instrument is given by the relation

$$N = \int_{\Delta\nu} N_{\nu}^0(T) [1 - \exp\{-k(\nu, T)u\}] \tau(\nu) d\nu \quad (3)$$

where $\tau(\nu)$ is the spectral transmission function of the instrument and the integral is taken over all frequencies to which the instrument responds at a given spectral setting. In gaseous emitters $k(\nu)$ can vary by large amounts over a narrow spectral range as one passes from line to line in the spectrum. The non-linear dependence of the emissivity on the absorption coefficient often results in great deviations of the measured emissivities from the Beer-Lambert Law. For this reason, considerable care must be taken in interpolating or extrapolating gaseous emission to optical depths other than those at which measurements are available.

In certain situations, and for specific ends, the use of measured spectra to compute ϵ and N at different optical depths is permissible. Use of Eq. (1) will always provide an upper limit to the emission from gas samples at lengths greater than those of the measured sample. Furthermore, if the measured gas is optically thin, the emission in various spectral regions (whose width is greater than the resolving power of the spectrometer) from gas samples of lesser optical depths than the measured

sample may be calculated exactly. The requirement for optical thinness is that the peak value of ku within the spectral interval under consideration be small with respect to unity. For practical purposes, a gas is optically thin if the value of the integrated intensity^{*} of a vibration-rotation band, determined from the measured spectrum, is equal to the value obtained in the limit as u is made very small.

Another situation pertains when the pressure or the temperature of the gas is high. At high pressures, the energy levels of molecules are perturbed by the close proximity of other molecules during collisions. As a result,² the spectral line emitted because of a particular class of transitions is spread over a wider frequency interval and less intense at the peak than at low pressures. At high temperatures, energy levels are excited which are not excited at low temperatures. Transitions involving these energy levels often give rise to emission at frequencies between the peaks of low-temperature lines. This filling in of the "valleys" in the spectrum which occurs at high pressures and temperatures

* The integrated intensity of a band is defined by the relation

$$\alpha_b = \int_{\Delta\nu} k_b(\nu, T) d\nu$$

where k_b is the contribution of the band in question to the absorption coefficient. The value of α_b is related to the quantum-mechanical probability of spontaneous transition from an upper to a lower energy level.

results in the spectral absorption coefficient's being a much less jagged function of frequency and consequently to the satisfaction of Eq. (1) over reasonably wide spectral intervals. The nearness of the integrated band intensities of the measured emission spectrum to the true values at very low pathlength can again be used as a test for this behavior. Under conditions of very high pressure, or in solids or liquids, the emission at great optical depths is given exactly by an extrapolation of the results of measured data by means of Eq. (1). More generally, even though the pressure or temperature is high enough to make the measured integrated band intensity the correct value, only extrapolations to lower optical depths are exact; however, the calculations for greater optical depths do again provide an upper limit to the emission.

One is normally interested in the emission from hot water vapor at the temperatures produced in combustion systems - flames, furnaces, engines, and rocket exhausts - which may range between 500° and 2500°K . At these temperatures most of the energy radiated by a blackbody falls at wavelengths longer than 0.9 microns, or at wavenumbers below $11,000\text{ cm}^{-1}$. (See Fig. 1) At low or moderate optical depths, emission from water vapor tends to fall proportionately more at longer wavelengths than does emission from a blackbody at the same temperature, and as the optical depth increases, the H_2O emission approaches that of a blackbody. We therefore conclude that we may replace the upper limit of integration in Eq. (2) by $11,000\text{ cm}^{-1}$ to a very good approximation, thereby avoiding having to consider the very high-order emission bands of water below $0.9\text{ }\mu$. The important bands of H_2O in this region are the pure rotational

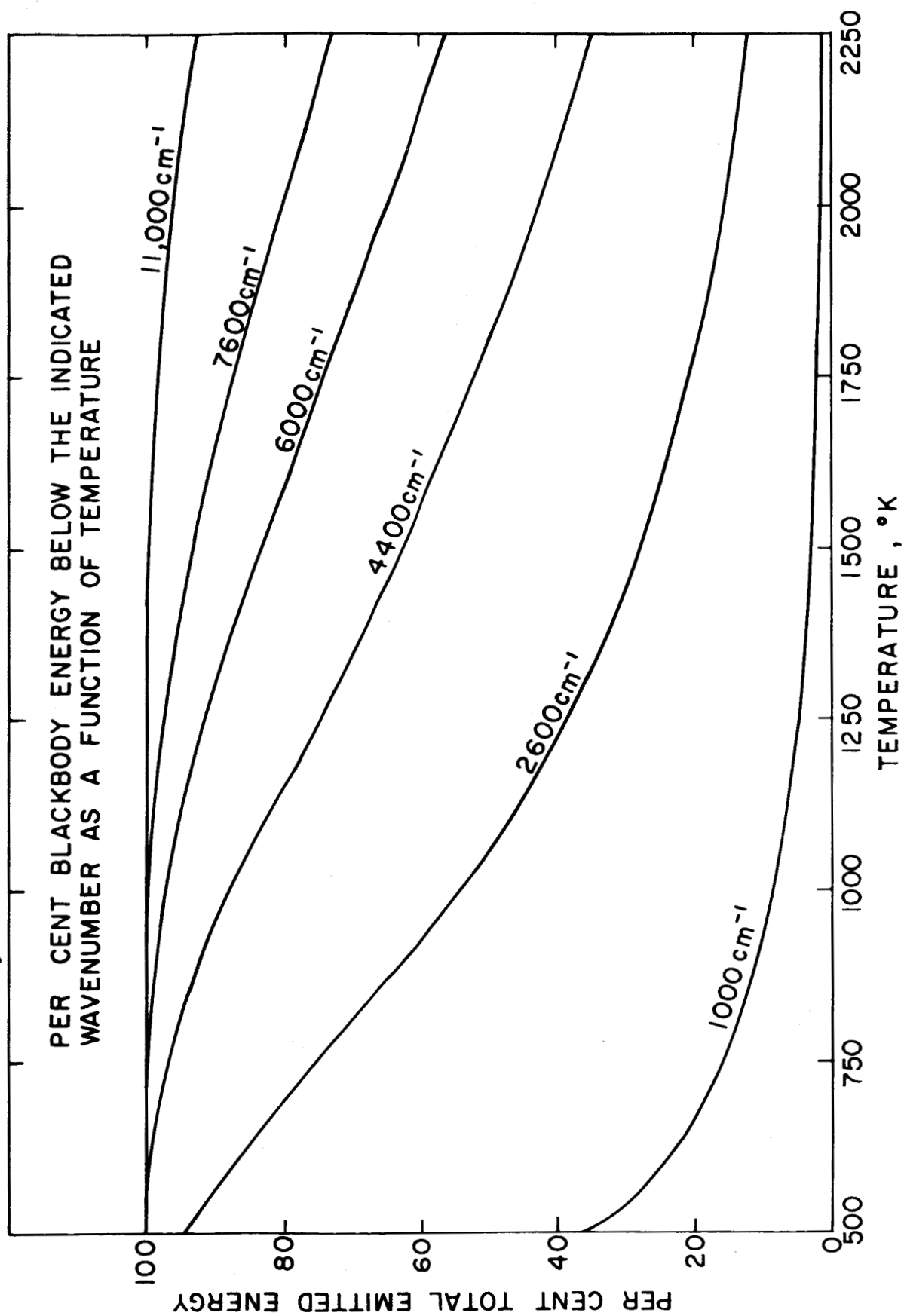


Figure 1. Blackbody Spectral Energy Distribution

band, the 6.3- μ vibration-rotation fundamental, the 2.7- μ fundamental, the 1.87- μ vibration-rotation overtone-combination, the 1.34- μ overtone, and the 1.14- μ overtone.

TREATMENT OF EXPERIMENTAL DATA

Spectral emissivities of the vibration-rotation bands and of the high-frequency side of the pure rotation band have been measured by Ferriso and Ludwig²⁻⁶ at a total pressure of one atm, temperatures between 530° and 2200°K, and at optical depths between 0.8 and 2.34 cm·atm of H₂O. The integrated intensities of the vibration-rotation bands, at temperatures greater than 1500°K, determined from these data and corrected for the temperature dependence of overtone and combination bands, generally agree well with the data obtained by other investigators⁷⁻¹⁰ using quite different techniques. The present data were measured in gases produced at the exit of a small rocket motor with a contoured nozzle designed to produce a uniform exhaust. Details of the experimental technique may be found in previous publications.²⁻⁵ Typical results (synthesized from measurements of the various band systems) are shown in Fig. 2, where the spectral emissivity of the exhaust gases is plotted versus wavenumber.

From the measured emissivity values, one may determine an apparent absorption coefficient, Z , by inverting Eq. (1):

$$Z(\nu, T) = \frac{1}{u} \ln \frac{1}{1-\epsilon} \quad (4)$$

At high temperatures, Z approaches \bar{k} , the "true" absorption coefficient

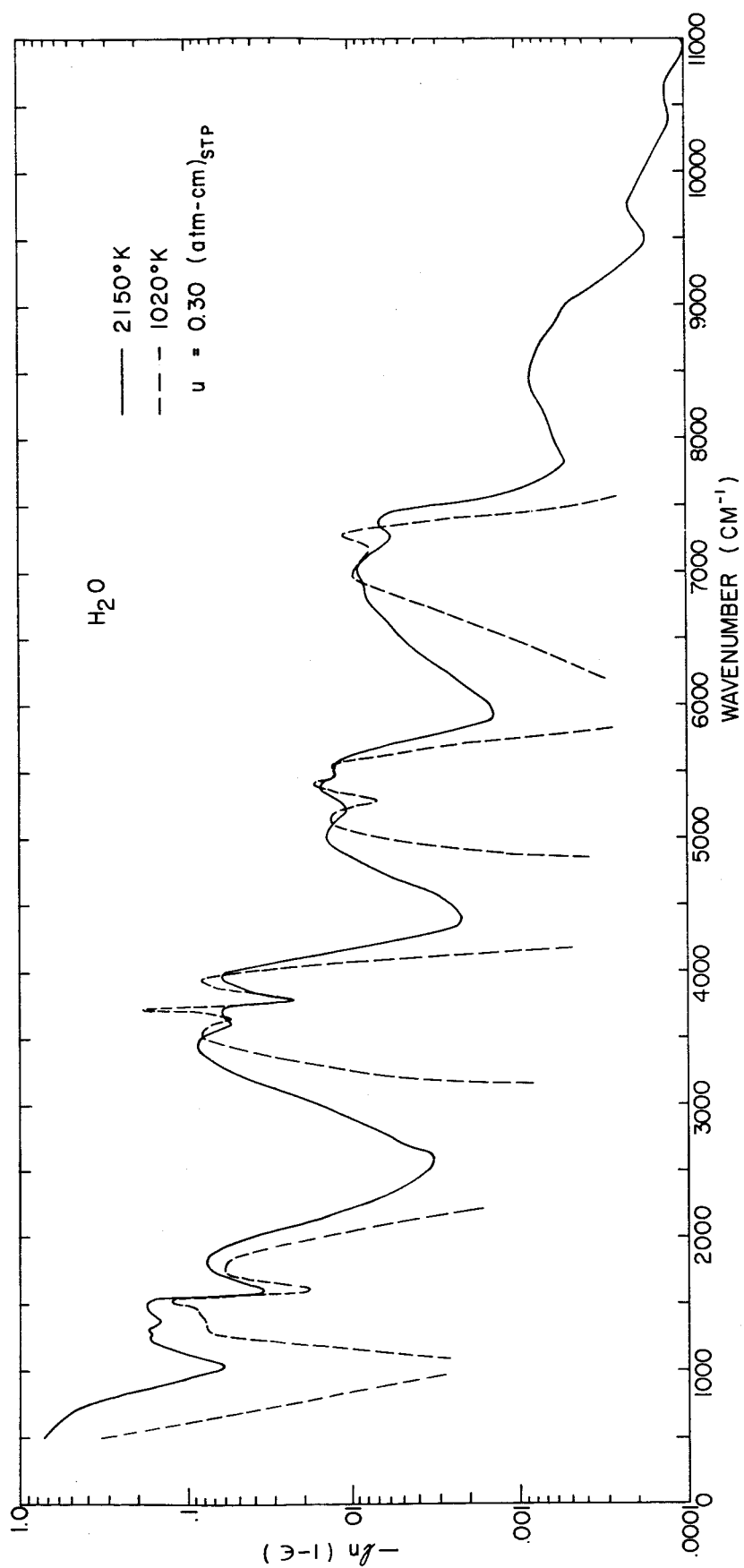


Figure 2. Spectral Emissivity of H_2O

frequency-averaged over a small interval. This may be seen from Table I, where the STP values of the apparent integrated intensities of the vibration-rotation bands (corrected for the theoretical temperature dependence in the case of the overtone bands) are tabulated at various temperatures. Constant values, comparable to other measured values, are attained at high temperatures. At lower temperatures, appreciable self-absorption occurs, so that $Z \neq \bar{k}$. The aim of the present investigation is the determination of values of the total emissivity of water vapor which are exact at low pathlengths and which represent a maximum upper limit at moderate and long paths. We therefore may use the high-temperature values directly in Eq. (2) to determine emission from high-temperature water, but the low-temperature values require correction.

In the case of the vibration-rotation bands, the correction is made by use of the expression for the spectral emissivity of a gas represented by the statistical¹² band model with collision-broadened lines. In this case

$$Z = \bar{k} / \sqrt{1 + \bar{k}u/4a}, \quad (5)$$

where a is the ratio of the line half-width to the mean line spacing. Here both \bar{k} and a are functions of temperature, wavelength, and partial pressures of the various constituents of the gas. In principle, the determination of good values of \bar{k} and a from measured data requires measurements on samples of at least two different pathlengths at the

TABLE I

APPARENT INTEGRATED INTENSITIES OF H₂O VIBRATION-
ROTATION BANDS VS TEMPERATURE

T	$\alpha_{6.3}$	$\alpha_{2.7}$	$\alpha_{1.87}$	$\alpha_{1.34}$	$\alpha_{1.14}$
500	150	150	-	-	-
1000	160	155	29	19.5	-
1200	210	170	-	19.5	-
1500	280	180	30	20	-
1800	310	190	28.5	22	-
2200	320	180	27	21.5	1.92
"best value"*	300	180	26	20	0.9

*From our data and those of other investigators.

same temperature, partial pressures, and wavelength. (This is the purpose of the present NASA-funded study using the long burner.) At present, however, such data are not available, and another approach is required. Here the values of a are averaged over the band to provide an "effective" band value, \bar{a} . The values of \bar{k}_{app} at small wavenumber intervals and of \bar{a} for the band are then determined from Eq. (5) and the condition that the integrated band intensity be equal to that determined from the high-temperature data, i.e.,

$$\int_{\text{band}} \bar{k}_{app}(T, \nu) d\nu = \alpha \quad (6)$$

Because of the use of a band-averaged \bar{a} , \bar{k}_{app} is not truly \bar{k} . However, since \bar{k}_{app} was determined in part from an integral relation, Eq. (6), its use in another integral relation, Eq. (2), should not lead to large errors provided that N_{ν}^0 , the blackbody function, varies by less than an order of magnitude over the band. This procedure has been applied to the 6.3- μ and 2.7- μ bands of H_2O ; correction of the overtone and combination bands was unnecessary. Values of $\bar{a}(T)$ below 1500°K for the various bands are close to those expected for H_2O from theoretical considerations.¹³ At temperatures above 1500°K, good values of \bar{a} cannot be determined from the data at hand, but they are not required for these cases.

In the case of the pure rotation band, only the high-wavenumber side of the band has been measured. Moreover, the integrated band intensity cannot be simply related to a vibrational transition probability

and therefore has a complex temperature dependence. A somewhat different approach has been used here, namely that of specifying the values of \bar{k} by an analytical function which is determined from theoretical considerations. The calculation is complicated by the fact that the H_2O molecule is asymmetric, so that no simple expression for the rotational energy levels exists. However, analytical expressions have been developed for the case of a prolate spheroid by Penner¹ and for the "most asymmetric molecule" by us.⁶ Penner's calculation can be expressed as*

$$\bar{k}_v^{PS} = \frac{8\pi^3}{3hc} \mu_o^2 N_o \frac{273.1}{T} \frac{\sqrt{(1+\beta)/\beta}}{2\gamma} u^2 e^{-\frac{u^2}{4\gamma}} \text{erf}\left(\frac{\sqrt{\beta}u}{\sqrt{4\gamma}}\right) [1-e^{-u}], \quad (6)$$

where $u = h\nu/kT$, $\gamma = hc\sqrt{BC}/kT$, $\beta = (A/\sqrt{BC}-1)$, μ_o is the permanent dipole moment of H_2O , N_o the particle density at STP, and A, B and C are the three rotational constants of H_2O . For water, $\mu_o = 1.87$ debyes, $\beta = 1.482$, and $\sqrt{BC} = 11.6 \text{ cm}^{-1}$. The expression for the "most asymmetric molecule" is

$$\bar{k}_v^{AS} = \frac{8\pi^3}{3hc} \mu_o^2 N_o \frac{273.1}{T} \frac{u}{\gamma^{*2}} \sinh\left(\frac{u^2}{8\gamma^*}\right) e^{-\frac{u^2}{4\gamma^*}} e^{-\frac{u}{2}} [1-e^{-u}] \times \frac{\gamma^{3/2} \sqrt{1+\beta}}{\sqrt{\pi}} \quad (7)$$

where $\gamma^* = \frac{1}{2}(A+C) hc/kT$ and other symbols have the same meaning as in Penner's expression. Both expressions assume just-overlapping rotational

* Penner omits the last term, which accounts for induced emission and is very important at low frequencies.

lines. We have found that where we can separate the emission due to the rotational band from that due to the 6.3- μ band, the emissivity in the wing of the rotational band is rather well described by setting

$$\bar{k} = K \bar{k}^{\text{PS}} + (1-K) \bar{k}^{\text{AS}}, \quad (8)$$

where K is the asymmetry parameter. Farther into the rotational band, the emission is overestimated by an amount corresponding to a statistical-model value of \bar{a} which does not appear unreasonable. We have therefore chosen to represent the absorption coefficient of the rotational band by Eq. (8). Emission from the pure rotation band is a significant fraction of the total at low temperatures and at very short pathlengths at high temperatures; we estimate a maximum-possible error of ± 20 per cent for the emission in the rotational band due solely to our use of the analytical representation for \bar{k} .

RESULTS

We have evaluated Eq. (2) over the range of wavenumbers from 0 to 11,000 cm^{-1} , using data prepared in the manner described above. The integration was performed at various optical depths by means of the CDC 160A computer. The results may be expressed in a number of ways. One may define the total emissivity by the relation

$$\epsilon_T(u, T) = N(u, T) / \sigma T^4, \quad (9)$$

where σ is the Stefan-Boltzmann constant. In Fig. 3 we show the total emissivity (in our upper-limit approximation) of hot water vapor as a function of temperature for parametric values of the optical depth u . (The solid curves were calculated; the dotted curves were graphically interpolated from plots of ϵ_T vs u .)

The frequency distribution of the emission from water vapor changes considerably with changes in temperature and pathlength. In Fig. 4 we show the emission per cm·atm optical depth at very thin paths from water vapor at 500°K; here approximately half of the emission is due to the rotational band, and the 2.7- μ band is relatively unimportant. In Fig. 5 we show the emission for a pathlength of 100 cm·atm of water at 500°K; here the 6.3- μ band contributes over two-thirds of the emission, though the rotational band is still important. In Fig. 6 we show the emission per cm·atm from thin water vapor samples at 2200°K; the 2.7- μ band assumes considerable importance and the higher-order 1.87- and 1.34- μ bands appear. Finally, in Fig. 7, we show the emission from 100 cm·atm of water at 2200°K; the rotational band is relatively unimportant, the 2.7- μ band dominates, and the higher-order bands contribute about one-fourth of the total emission. The changing character of water emission at different temperatures and optical depths is well-illustrated by these graphs.

The fractional contribution from the spectral region below a given wavenumber ν_f to the total emission can be calculated as

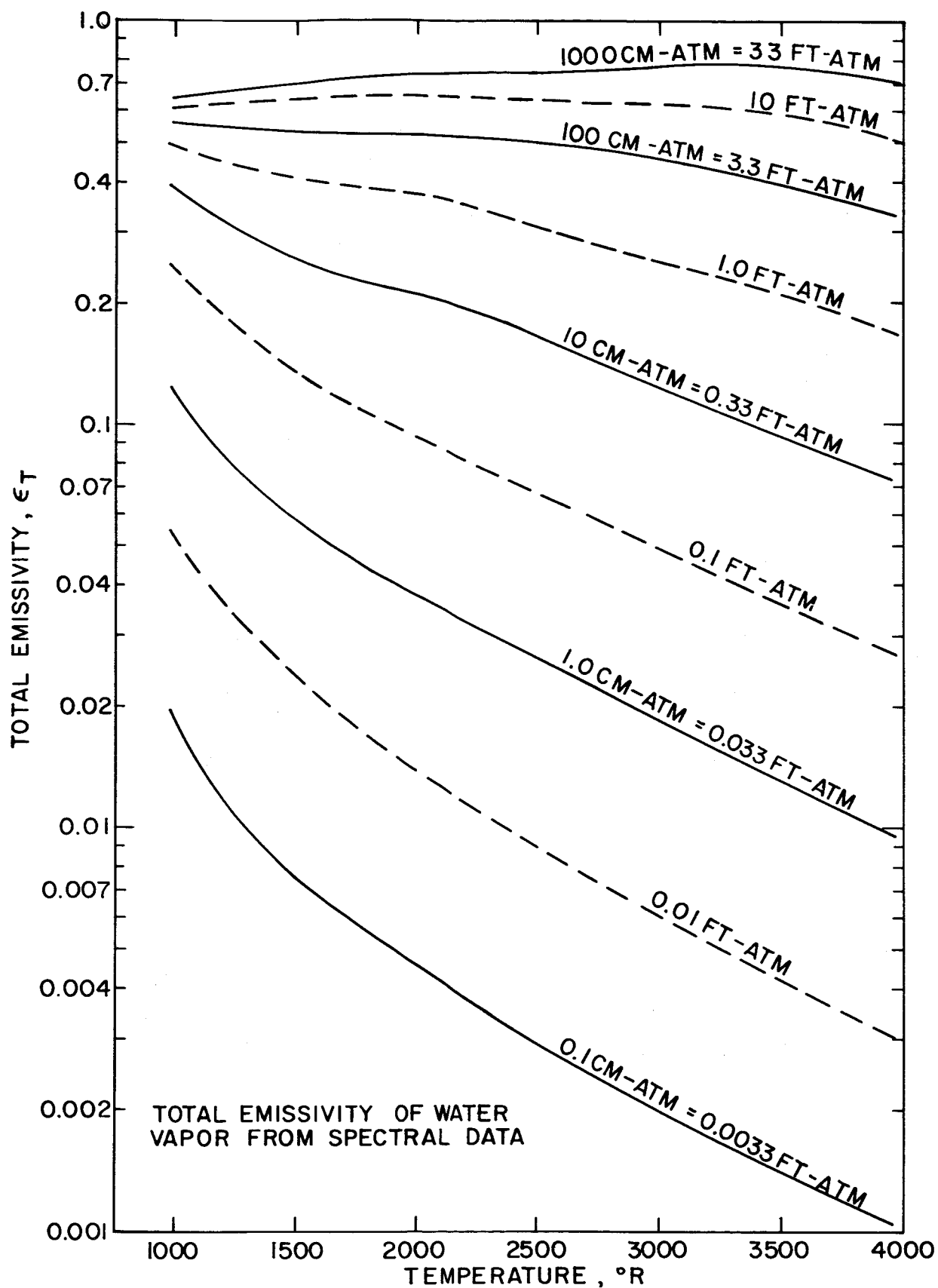


Figure 3. Total Emissivity of H_2O Vs. Temperature

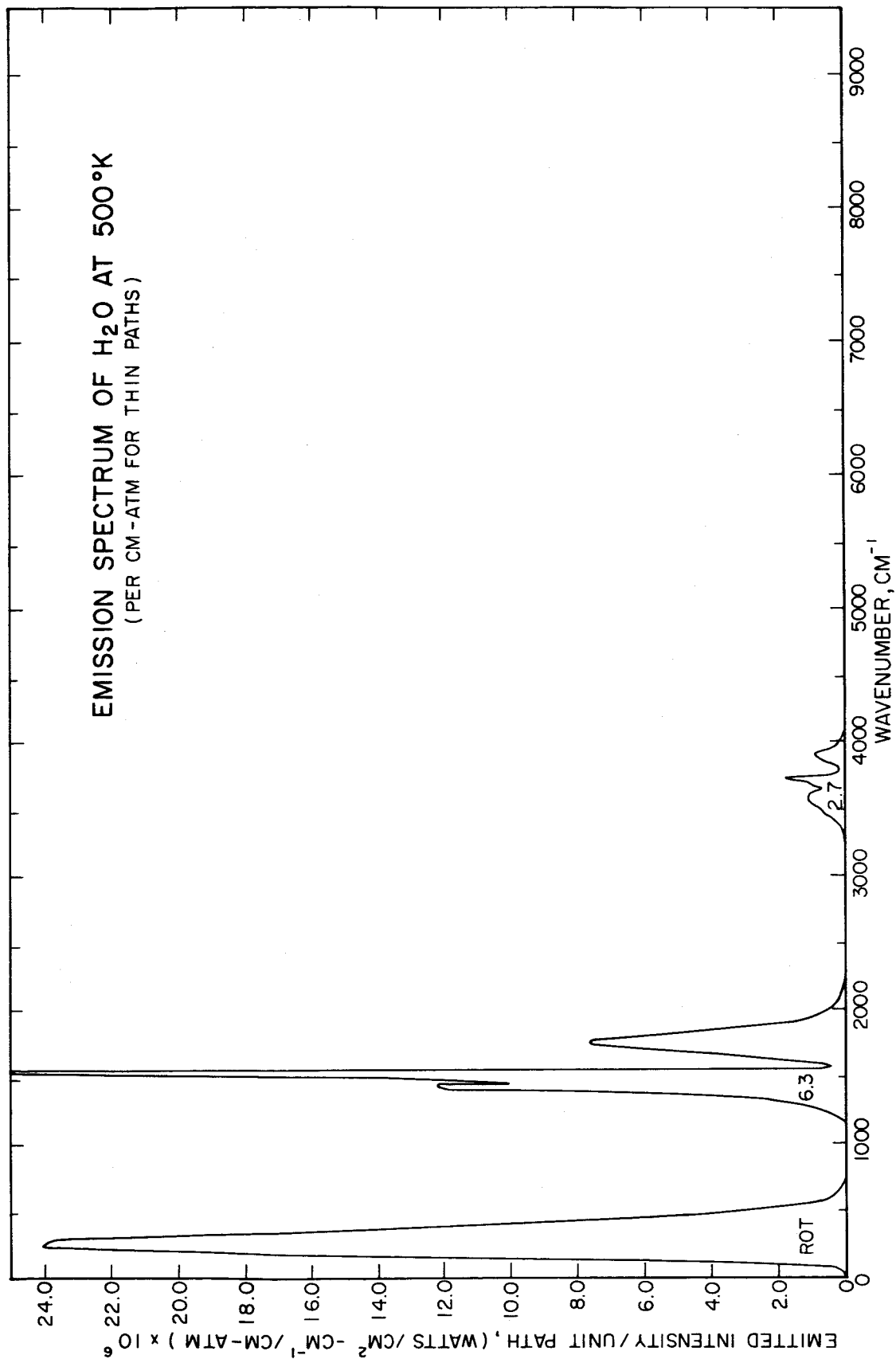


Figure 4. Spectral Radiant Emission from H₂O at 500°K and Very Small Optical Depth

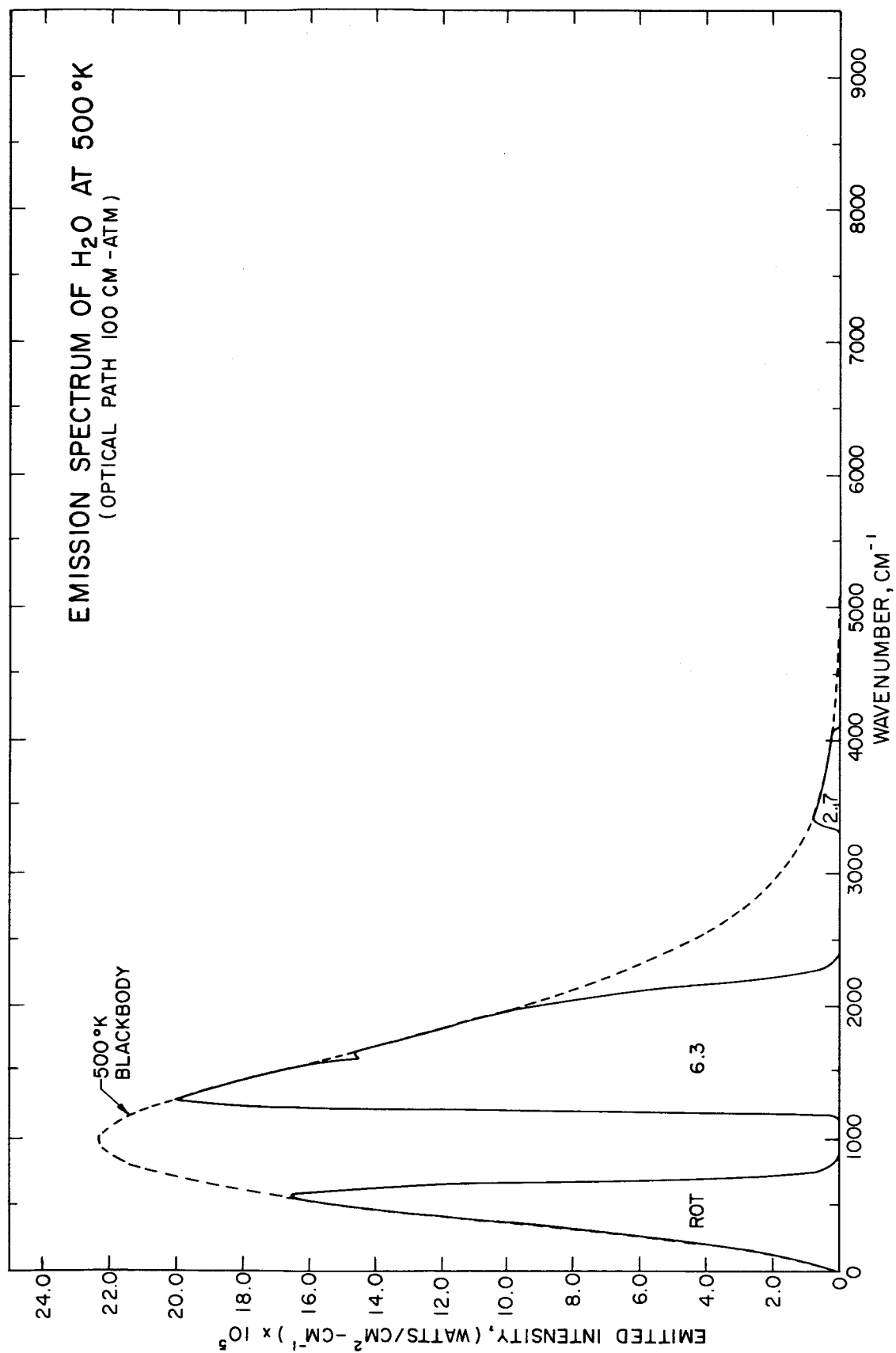


Figure 5. Spectral Radiant Emission from H₂O at 500°K and 100 cm·atm Optical Depth

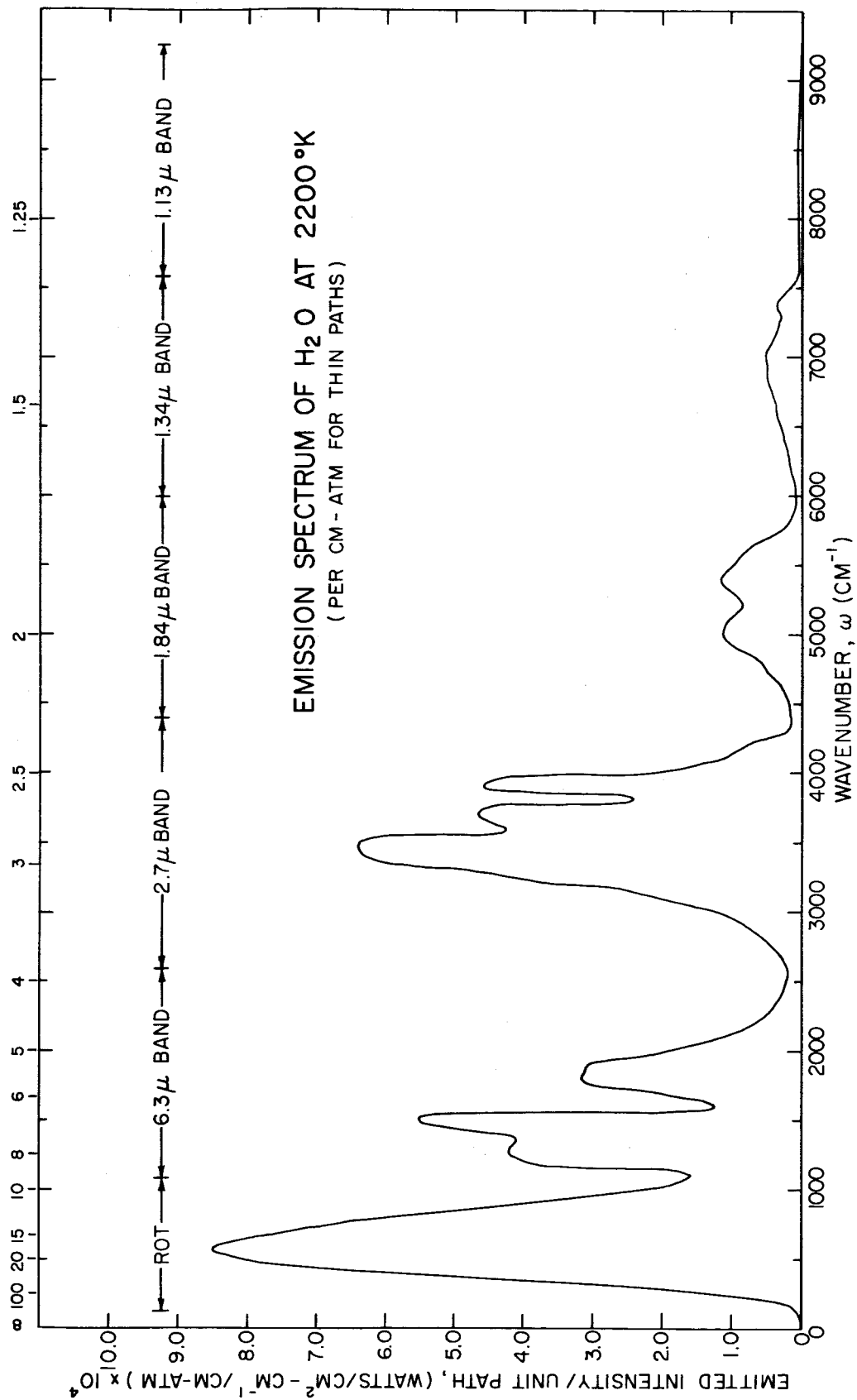


Figure 6. Spectral Radiant Emission from H_2O at $2200^\circ K$ and Very Small Optical Depth

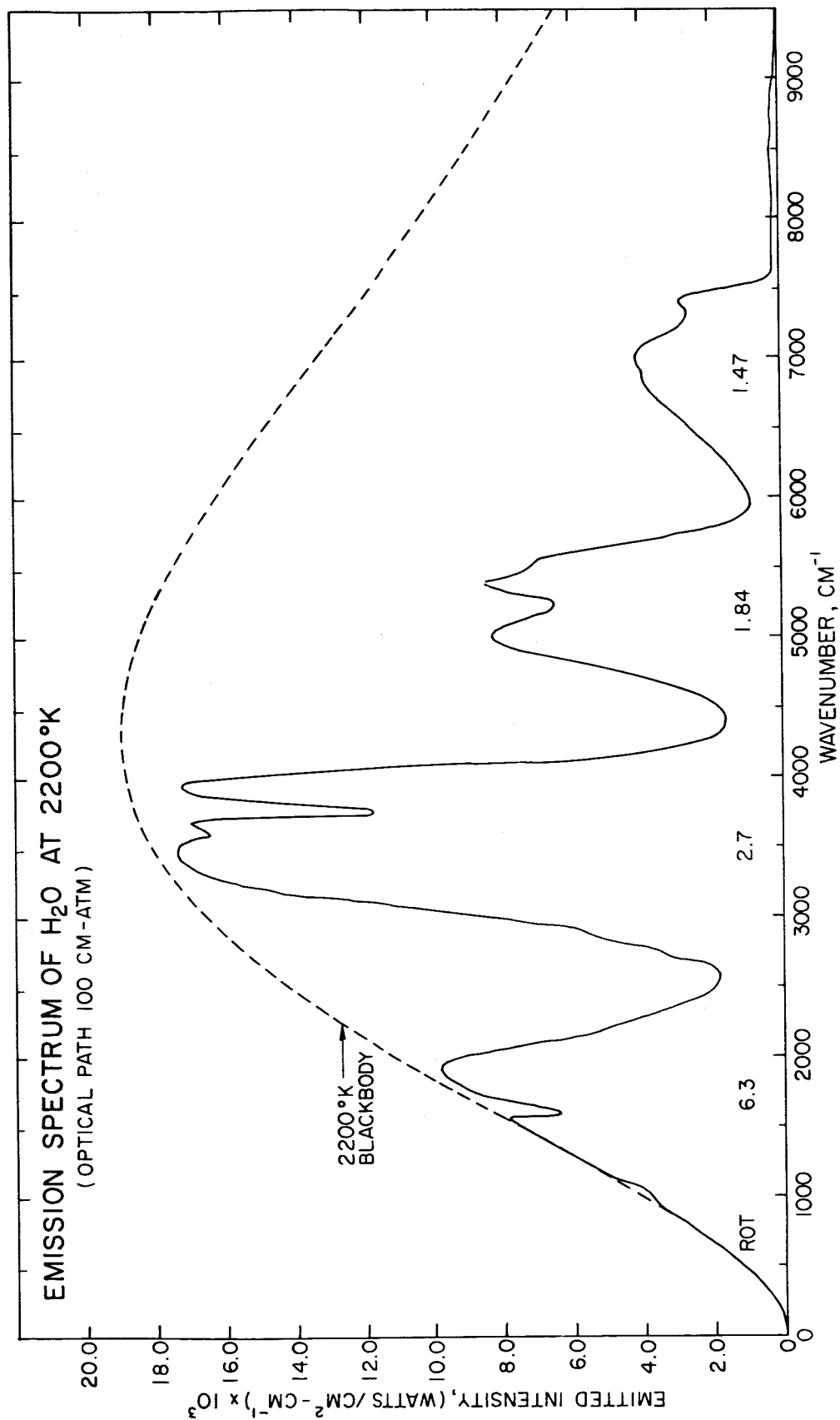


Figure 7. Spectral Radiant Emission from H₂O at 2200°K and 100 cm·atm Optical Depth

$$f_j(u,T) = \int_0^{\nu_j} N_{\nu}^o(T) \epsilon(\nu,T,u) d\nu / N(T,u) . \quad (10)$$

These data are shown in Figs. 8, 9, and 10 for optical depths of 0.1, 10, and 1000 cm·atm. The dividing points have been chosen so that the different spectral regions correspond approximately to the various emission bands of water vapor. The correspondence is not quite exact at the highest temperatures, since there is some overlapping of the bands under these conditions. These data may be compared with the blackbody distribution shown in Fig. 1 to see the differences caused by the non-graybody behavior of water vapor under various conditions.

COMPARISON WITH PREVIOUS WORK

The data presented in Fig. 3 can be directly compared with those of Hottel,¹⁴ which are generally regarded as the best total emissivity data extant up to now. Replottings of our data and Hottel's data are shown in Figs. 11 and 12. At large optical depths, we find that under conditions where Hottel has actual measurements the present values of total emissivity are higher than his. One expects this behavior at long paths, since our calculations give an upper limit to the emissivity here. It is somewhat surprising that the present values are also considerably higher than Hottel's when the optical depth is small. Under these conditions our values should be exact, subject only to the high-frequency cutoff on limits of integration and to uncertainties in the true values of the absorption coefficient of the pure rotation band.

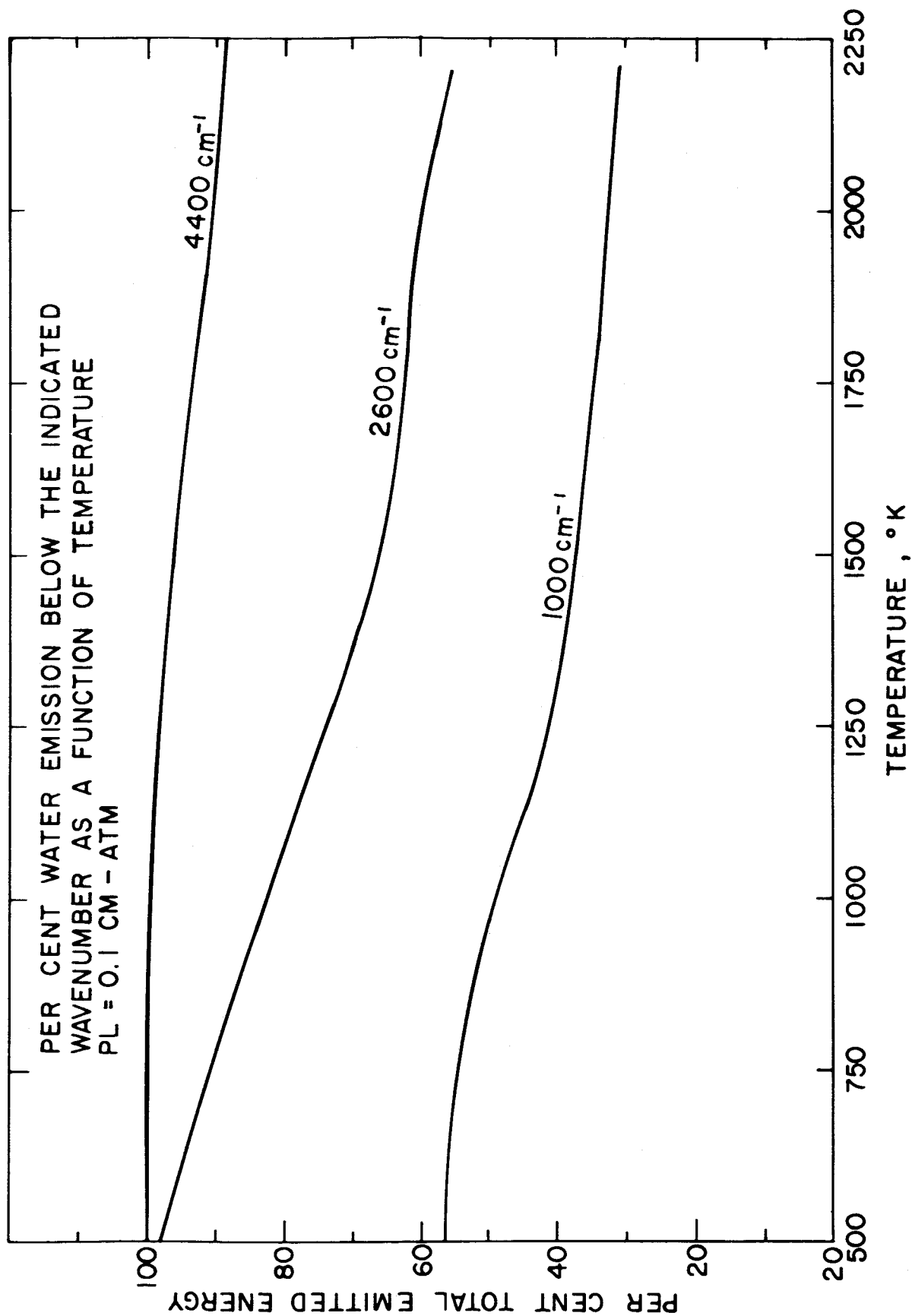


Figure 8. Spectral Energy Distribution of 0.1 cm·atm of H_2O

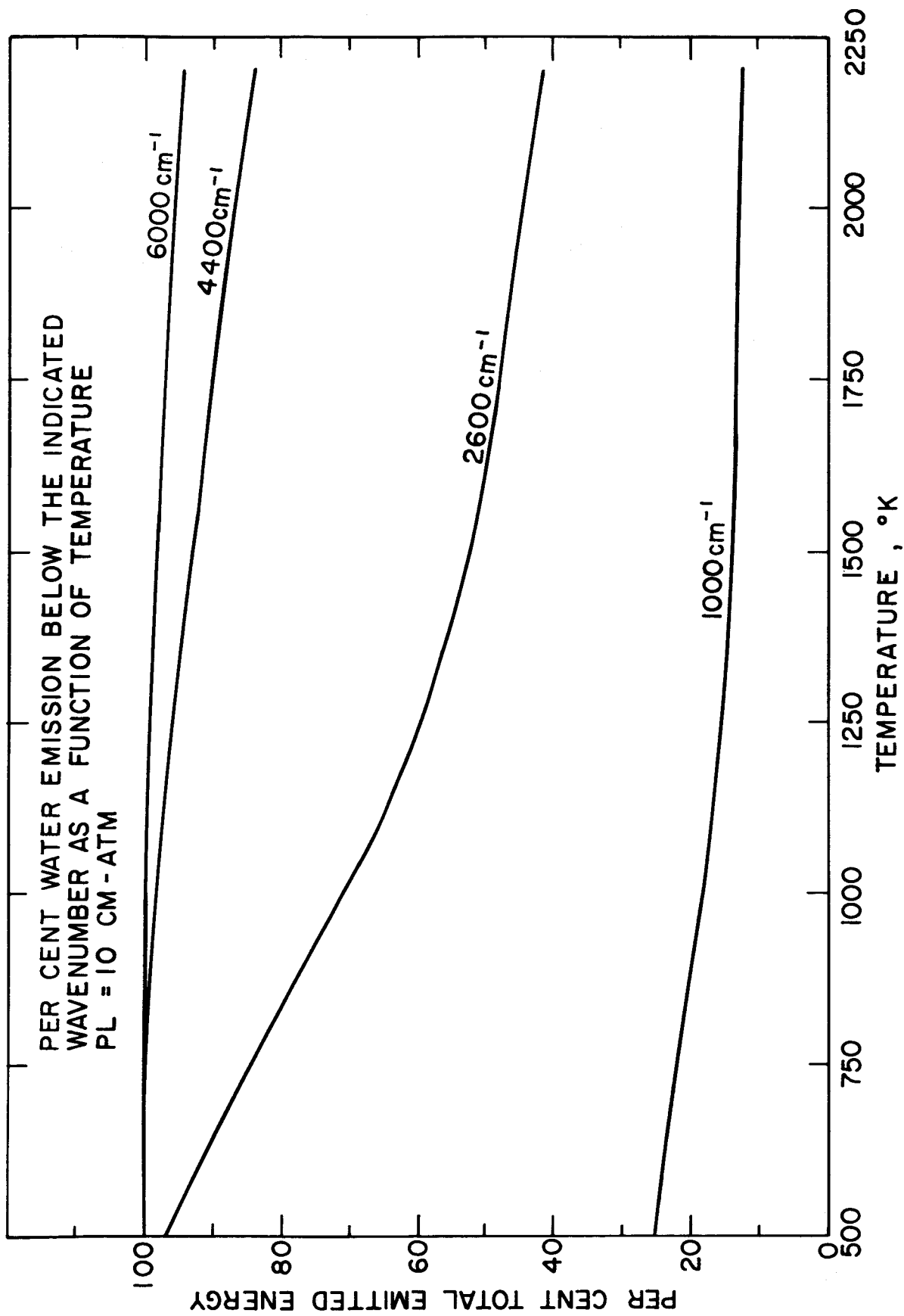


Figure 9. Spectral Energy Distribution for 10 cm³/atm of H₂O

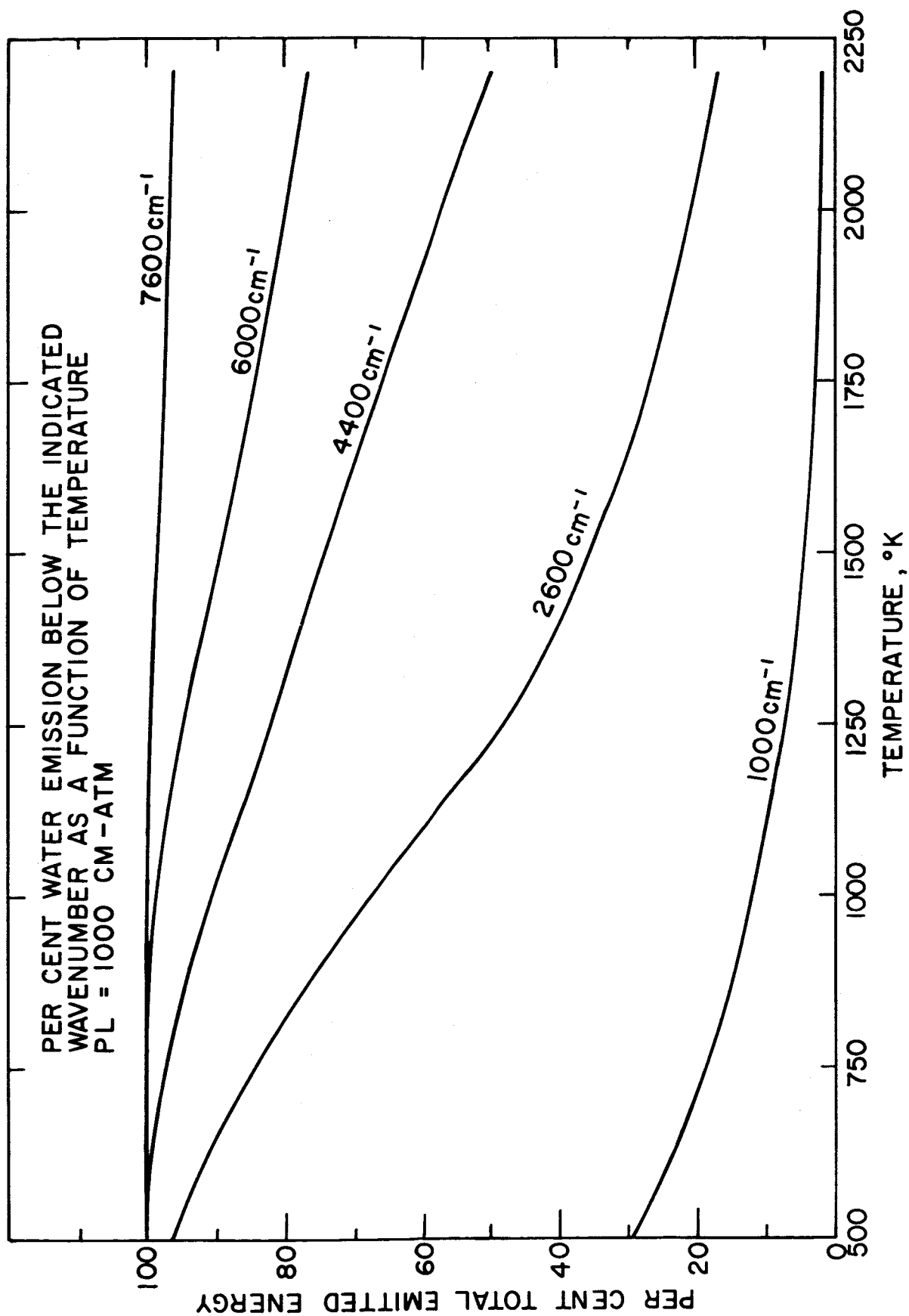


Figure 10. Spectral Energy Distribution for 1000 cm·atm of H_2O

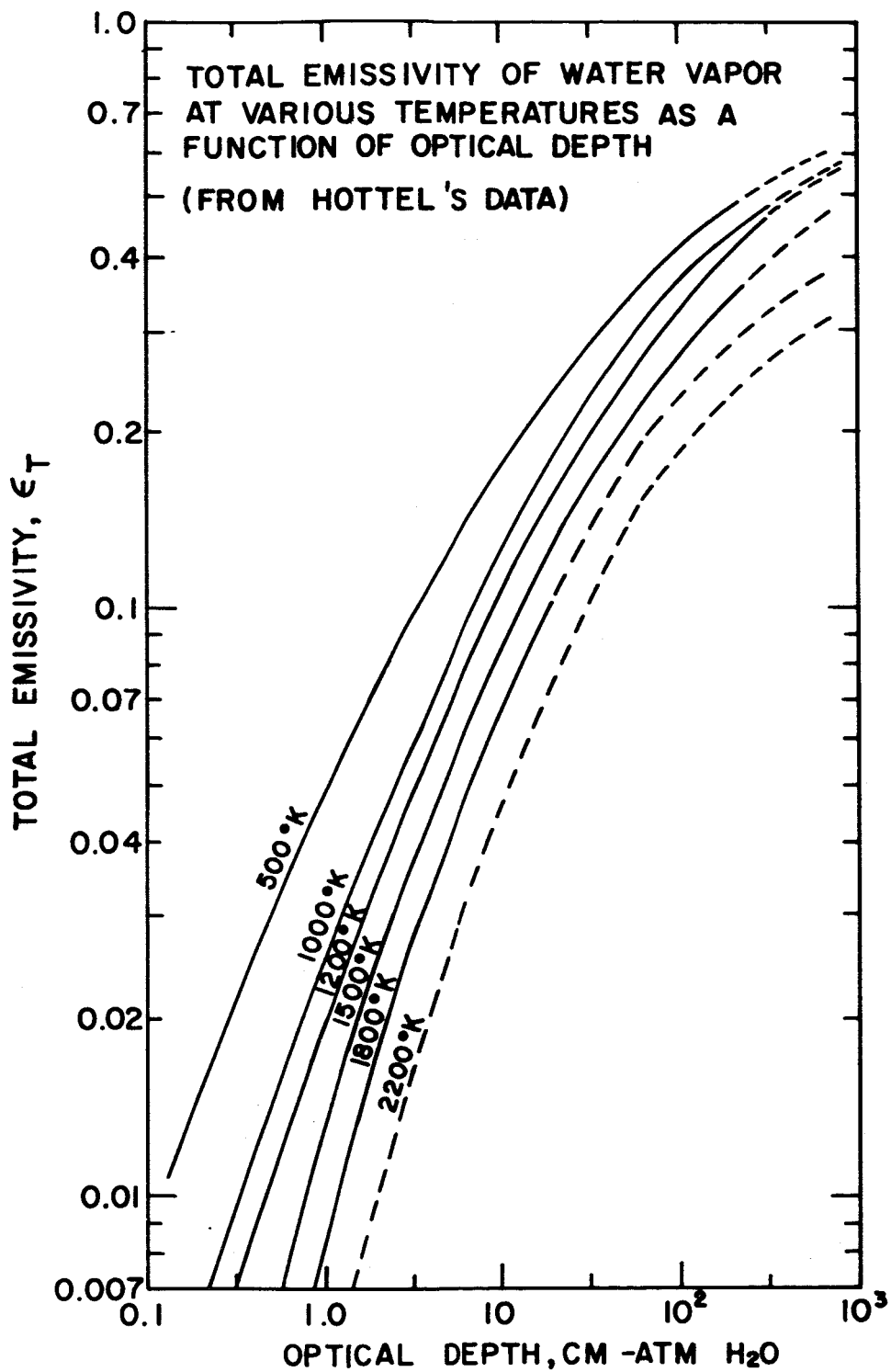


Figure 11. Total Emissivity of H_2O (from Hottel) Vs. Optical Depth

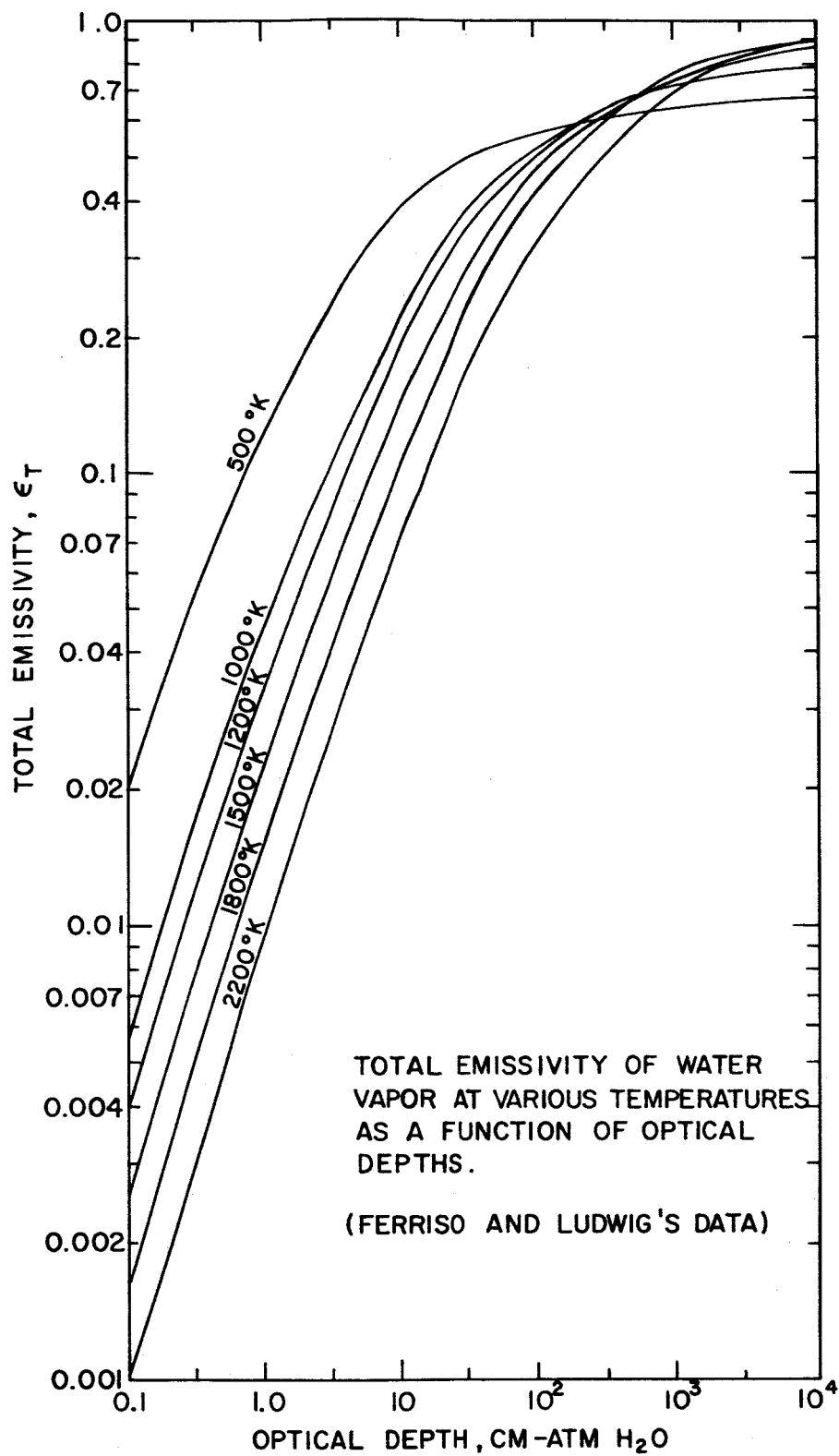


Figure 12. Total Emissivity of H_2O (from Ferriso and Ludwig) Vs. Optical Depth

The first error actually reduces our values, while the second should not cause more than a 10 per cent overestimation in the worst case. There are several possible causes for the disagreement, which varies from a factor of 2.4 at 500°K to a factor of 1.5 at 2200°K.

First, we note that while the data we show here were obtained spectrally, with separate instrumental response calibrations at all observed frequencies, Hottel's were total measurements and his calibration was affected by exposing his detector to blackbody radiation of all frequencies at once. Variations in the spectral response of Hottel's detector, not shown by his calibration, may be responsible for the differences between our data and his. In at least one series of experiments¹⁵ he exposed his detector to blackbody radiation at temperatures of 660 and 1270°K, and found only a three per cent difference in the calibration constant, indicating that the response of his detector was most likely reasonably flat between 1000 and 5000 cm^{-1} . However, much emission at short paths (particularly at low temperatures) occurs below 1000 cm^{-1} .

A second possible source of error is in temperature measurement. If our measured temperatures were consistently low, or Hottel's consistently high, then the emissivities derived from these data and the measured gaseous emission would differ in the manner shown. Hottel made his measurements of temperature with thermocouples and by sodium-D line reversal. His thermocouple traverses¹⁶ show little temperature variations along the line of sight, and seem to have taken account of thermocouple radiation.

We have not enough information to judge the sodium measurements. In our system, we have measured the temperature by means of an infrared absorption-emission technique.¹⁷ We find that the temperatures we measure are very close to those measured in the same rocket motor by Griggs using a sodium reversal technique.¹⁸ Furthermore, the integrated band intensities derived from our measurements (at high temperatures) provide an independent check on the data. We find close agreement with the data of other investigators, which could not occur if our temperature measurements contained a consistent error.

Atmospheric absorption provides a third source of error. It is well known that radiation from a gas, which consists of discrete lines, is more strongly absorbed by a cool layer of the same gas, which absorbs in mostly the same lines, than is radiation from a spectrally continuous source. Both Hottel and ourselves recognized the importance of absorption and took pains to minimize it. In our case, the relative absorption at any wave-number was never greater than 5 per cent, and the absorption at most frequencies is truly negligible. Furthermore, our method of calibration tends to minimize this difference. If in spite of his precautions Hottel's experiments were performed with the presence of absorbing layers between the emitting gas and his detectors, then his measured emissivities would be low. (An absorbing layer of constant optical depth would affect his thin-gas data more than the optically thick data, and would be especially important to the very intense rotational band.)

We have not been able to pinpoint the source of the difference

between Hottel's optically thin H_2O emissivities and those we have calculated from our spectral measurements. Since our data have a built-in independent check in that integrated band intensities can be compared with those of other investigators, we feel that our data in this region are more likely to be correct. At the present time we cannot say whether the disagreement extends to larger optical depths, because the present calculations provide only an upper limit to the emissivity under those conditions.

SUMMARY

The total emissivity of gases for very thin optical paths can be calculated exactly and straightforwardly from measured gas spectral emissivity data provided that the observed gas is itself optically thin. An absolute upper limit to the total and spectral emissivity can be calculated by use of the thin-gas spectral absorption coefficients and Beer's Law. The experimental data of Ferriso and Ludwig for water vapor spectral emissivities satisfy the requirement of optical thinness at high temperatures, except for the pure rotational spectrum. Values of the spectral absorption coefficient frequency-averaged over a small wave-number interval can be derived from their low-temperature data for the 2.7- and 6.3- μ bands. The rotational band frequency-averaged absorption coefficients are calculated from approximate theoretical expressions which give good qualitative agreement with the measured data. The corrected and supplemented data of Ferriso and Ludwig have been employed in the

Beer's Law relation to obtain upper-limit spectral emission for H_2O at temperatures from 500° to 2200°K , and optical depths from 0.1 to 10,000 $\text{cm}\cdot\text{atm}$. The data thus generated are integrated over the wavenumber range from 0 to $11,000\text{ cm}^{-1}$ to obtain total emissivities of H_2O for the various conditions. Comparison of these data with those of Hottel shows that the total emissivities derived from Ferriso and Ludwig's data are considerably higher than Hottel's when the gas is very thin, where the present calculation is exact. We believe that the thin-gas data presented here are correct, but have not been able to ascertain precisely the cause of the discrepancy between our data and Hottel's.

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